

930,988



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## PATENT SPECIFICATION

NO DRAWINGS

930,988

Date of filing Complete Specification: June 26, 1959.

Application Date: July 3, 1958.

No. 21308/58.

Complete Specification Published: July 10, 1963.

Index at acceptance:—Classes 37, K(1D3A:2R4:3X), K4(F1:G4:GX); 2(3), C1F4(B:D3:F2:F5), C2B(18:21); 2(5), R1C(9:10:11:12:13:14:16), R2C(9:10:11:12:13:14:16), R3C(6:7:8:9:10:11:12:13:14:16), R5C(9:10:11:12:13:14:16), R19C(9:10:11:12:13:14:16), R20C(9:10:11:12:13:14:16), R22C(9:10:11:12:13:14:16), R29C(9:10:11:12:13:14:16), R33C(9:10:11:12:13:14:16); 2(6), P3C(7:14B:17:20B), P10C(6B:14B:20C), P10(D1A:T2E); and 95, A4R, B4X.

International Classification:—H01c. (B05. C07c, d. C08f, g. C09d).

### COMPLETE SPECIFICATION

### Improvements in and relating to Electrophotographic Reproduction Materials

#### ERRATA

SPECIFICATION No. 930,988

Page 1, Table: 1st column, 11th entry, *delete* "hyphen" *after* "3"

Page 8, lines 53 and 54, *for* "formaldehyde" *read* "formaldehyde"

Page 9, line 30, *for* "image" *read* "images"

THE PATENT OFFICE

20th August 1963

25 overall uniform electrostatic charge in the dark or under safe light conditions and when exposed to light through an original, say a drawing, the electrostatic charge is lost in the areas exposed to light but retained to a greater or lesser degree, depending upon the amount of light to which the material is exposed, in the dark parts of the original. A charge pattern is thus produced on the material which corresponds with the original (e.g. the drawing) through which the charged plate was exposed.

30 This charge pattern may be rendered visible by the application of a developer powder of the kind which may acquire a charge, triboelectrically, the said powder being attracted to and held by the charge pattern produced by exposure in amounts depending upon the strength of the charge pattern.

in the manufacture of electrophotographic materials should possess certain characteristics such as for instance, range of use, reliability, simplicity of handling, light sensitivity and keeping qualities and it is an object of the present invention to provide materials for use in electrophotographic methods in which the photoconductive insulating materials used in their manufacture shall have as many of the above referred to desirable characteristics as possible.

According to the present invention there is provided an electrophotographic material consisting of a conductive support and a photoconductive insulating layer adherent hereto, which layer comprises an organic compound of general formula

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# POTENT SPECIFICATION

NO DRAWINGS

930.988

Date of filing Complete Specification: June 26, 1959.

Application Date: July 3, 1958.

No. 21308/58.

Complete Specification Published: July 10, 1963.

**Index at acceptance:**—Classes 37, K(1D3A:2R4:3X), K4(F1:G4:GX); 2(3), C1F4(B:D3:F2:F5), C2B(18:21); 2(5), R1C(9:10:11:12:13:14:16), R2C(9:10:11:12:13:14:16), R3C(6:7:8:9:10:11:12:13:14:16), R5C(9:10:11:12:13:14:16), R19C(9:10:11:12:13:14:16), R20C(9:10:11:12:13:14:16), R22C(9:10:11:12:13:14:16), R29C(9:10:11:12:13:14:16), R33C(9:10:11:12:13:14:16); 2(6), P3C(7:14B:17:20B), P10C(6B:14B:20C), P10(D1A:T2E); and 95, A4R, B4X.

**International Classification:**—H01c. (B05. C07c, d. C08f, g. C09d).

## COMPLETE SPECIFICATION

### Improvements in and relating to Electrophotographic Reproduction Materials

We, OZALID COMPANY LIMITED, a British Company and RICHARD PENN ROYER, a British Subject, both of Langston Road, Loughton, Essex, (formerly of 62, London Wall, London, E.C.2), do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to materials for use in electrophotographic reproduction methods. Electrophotographic methods are now well known and usually make use of the photoconductive insulating properties of certain materials such as selenium or zinc oxide. Such materials are capable of holding an electrostatic charge in the dark or under safe light conditions and of losing such charge selectively in accordance with the amount of light falling thereon.

In one such known method a metal plate coated with a layer of selenium is given an overall uniform electrostatic charge in the dark or under safe light conditions and when exposed to light through an original, say a drawing, the electrostatic charge is lost in the areas exposed to light but retained to a greater or lesser degree, depending upon the amount of light to which the material is exposed, in the dark parts of the original. A charge pattern is thus produced on the material which corresponds with the original (e.g. the drawing) through which the charged plate was exposed.

This charge pattern may be rendered visible by the application of a developer powder of the kind which may acquire a charge, triboelectrically, the said powder being attracted to and held by the charge pattern produced by exposure in amounts depending upon the strength of the charge pattern.

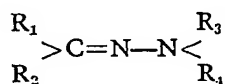
The visible powder pattern may now be transferred to a plain sheet of paper and fixed thereon, suitably by heat, to produce a copy of the original to which the material was exposed.

As mentioned previously selenium and zinc oxide are known to possess photoconductive insulating properties. Certain other materials, such as for instance sulphur, anthracene and anthraquinone have already been proposed for use in electrophotographic methods.

In the production of photoconductive insulating materials or so-called electrophotographic or xerographic plates the photoconductive material may be applied to a metal or other conductive base material either by vacuum deposition or as dispersion in solvents with a bonding agent such as a thermoplastic synthetic resin.

In view of the increasing importance of electrophotographic methods it is desirable that the photoconductive insulating materials used in the manufacture of electrophotographic materials should possess certain characteristics such as for instance, range of use, reliability, simplicity of handling, light sensitivity and keeping qualities and it is an object of the present invention to provide materials for use in electrophotographic methods in which the photoconductive insulating materials used in their manufacture shall have as many of the above referred to desirable characteristics as possible.

According to the present invention there is provided an electrophotographic material consisting of a conductive support and a photoconductive insulating layer adherent hereto, which layer comprises an organic compound of general formula



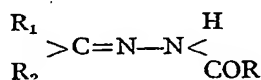
- in which  $R_1, R_2$  represent hydrogen, alkyl, aralkyl, acyl, aroyl, cycloalkyl, or univalent radicals of aromatic or substituted aromatic carbocyclic or heterocyclic ring systems and wherein  $R_1$  and  $R_2$  may join to form a ring, and in which  $R_3, R_4$  represent hydrogen, alkyl, aralkyl, aroyl, cycloalkyl, or univalent radicals of aromatic or substituted aromatic carbocyclic or heterocyclic ring systems and wherein  $R_3$  or  $R_4$  may represent acyl.

- Examples of such compounds include hydrazones of aldehydes such as the phenylhydrazones, the acylhydrazones, the benzaldehyde acetylhydrazones and the 9-anthraldehyde hydrazones.

- Of the phenylhydrazone group such compounds as benzaldehyde phenylhydrazone, benzaldehyde methylphenyl hydrazone, cinnamaldehyde phenylhydrazone, *p*-dimethyl aminobenzaldehyde, *p*-bromophenylhydrazone piperonal, *p*-bromophenylhydrazone, phenylacetaldehyde diphenylhydrazone, acetophenone phenylhydrazone, *o*-sulphobenzaldehyde phenylhydrazone and isonicotinic aldehyde phenylhydrazone may be used.

- Of the benzaldehyde acetylhydrazone group, benzylidene acetylhydrazide may be mentioned as an example.

- Many compounds of the acylhydrazone group having the general formula

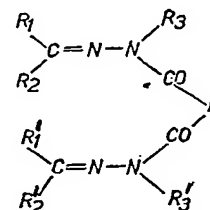


in which R represents H, alkyl, aralkyl, aryl, substituted aryl or a heterocyclic residue of

aromatic character,  $R_1$  represents H, alkyl, aryl or substituted aryl,  $R_2$  represents aralkyl, aryl, substituted aryl or a heterocyclic residue of aromatic character, and in which  $R_1$  and  $R_2$  may join to form a ring, may be used.

Compounds of this group may be prepared by condensation of equimolecular quantities of a carboxylic acid hydrazide with a compound containing a carbonyl function (e.g. aldehyde ketone, quinone) by boiling in an organic solvent, preferably ethanol.

The organic compound may also be formed by the reaction of a dihydrazide with an aldehyde or ketone to give a compound having the general formula:—



in which  $R_1, R_2, R_1^1, R_2^1$  represent hydrogen alkyl, aralkyl, acyl, aroyl, cycloalkyl, or univalent radicals of aromatic or substituted aromatic carbocyclic or heterocyclic ring systems wherein  $R_1, R_2$  and/or  $R_1^1, R_2^1$  may join to form a ring, in which R is alkylene and in which  $R_3, R_3^1$  represent hydrogen, alkyl, aralkyl, aroyl, cycloalkyl, or univalent radicals of aromatic or substituted aromatic carbocyclic or heterocyclic ring systems.

The following table gives in column 1 the starting carboxylic acid hydrazide, column 2 the reaction component with CO group content, the melting point of the acylhydrazone is given in column 3 and column 4 its colour.

1	2	3	4
benzhydrazide	4-dimethylamino-benzaldehyde	191 to 192° C.	pale yellow
benzhydrazide	4-diethylamino-benzaldehyde	180 to 181° C.	yellow
benzhydrazide	4-dibenzylamino-benzaldehyde	202 to 203° C.	yellow
benzhydrazide	anisaldehyde	157 to 158° C.	colourless
<i>o</i> -toluyl-hydrazide	4-dimethylamino-benzaldehyde	151 to 152° C.	yellow
4-methoxy-benzhydrazide	anisaldehyde	173 to 174° C.	colourless
4-methoxy-benzhydrazide	4-dimethylamino-benzaldehyde	230 to 231° C.	colourless

1	2	3	4
2-hydroxy-benzhydrazide	4-dimethylamino-benzaldehyde	264° C.	yellow
4-hydroxy-benzhydrazide	4-dimethylamino-benzaldehyde	238 to 239° C.	yellow
4-hydroxy-benzhydrazide	4-diethylamino-benzaldehyde	234 to 236° C.	yellow
2-amino-benzhydrazide	benzaldehyde	197 to 198° C.	colourless
2-methylamino-benzhydrazide	4-dimethylamino-benzaldehyde	218° C.	yellow
2-amino-benzhydrazide	4-dimethylamino-benzaldehyde	243 to 245° C.	yellow
N-methyl-di-phenylamine-4-carboxylic acid hydrazide	4-dimethylamino-benzaldehyde	200 to 201° C.	yellow
4-amino-benzhydrazide	4-dimethylamino-benzaldehyde	279 to 280° C.	yellow
diphenyl-4-carboxylic acid hydrazide	4-dimethylamino-benzaldehyde	257° C.	yellow
1-naphthoic acid hydrazide	4-dimethylamino-benzaldehyde	201 to 202° C.	colourless
2-amino-3-naphthoic acid hydrazide	4-dimethylamino benzaldehyde	247 to 248° C.	yellow
2-amino-3-naphthoic acid hydrazide	benzaldehyde	228 to 230°	yellow
2-hydroxy-3-naphthoic acid hydrazide	4-dimethylamino-benzaldehyde	243 to 244° C.	yellow
pyridine-4-carboxylic acid hydrazide	furfural	218 to 219° C.	yellow
pyridine-4-carboxylic acid hydrazide	9-ethyl-carbazole-3-aldehyde	247 to 248° C.	colourless
pyridine-4-carboxylic acid hydrazide	anisaldehyde	173 to 174° C.	colourless
pyridine-4-carboxylic acid hydrazide	piperonal	231 to 233° C.	colourless

1	2	3	4
pyridine-4-carboxylic acid hydrazide	4-dimethylamino-benzaldehyde	196 to 197° C.	yellow
pyridine-4-carboxylic acid hydrazide	2-chloro-4-dimethylamino-benzaldehyde	213 to 215° C.	yellow
pyridine-4-carboxylic acid hydrazide	4-diethylamino-benzaldehyde	191 to 192° C.	yellow
pyridine-4-carboxylic acid hydrazide	4-dibenzylamino-benzaldehyde	201 to 202° C.	yellow
pyridine-4-carboxylic acid hydrazide	1-naphthaldehyde	212 to 213° C.	yellow
pyridine-4-carboxylic acid hydrazide	anthracene-9-aldehyde	265 to 266° C.	yellow
pyridine-4-carboxylic acid hydrazide	anthraquinon-2-aldehyde	301 to 302° C.	yellow
pyridine-4-carboxylic acid hydrazide	cinnamaldehyde	201 to 202° C.	yellow
pyridine-4-carboxylic acid hydrazide	quinoline-4-aldehyde	195° C.	colourless
pyridine-3-carboxylic acid hydrazide	4-dimethylamino-benzaldehyde	146 to 147° C.	yellow
pyridine-3-carboxylic acid hydrazide	4-diethylamino-benzaldehyde	153 to 154° C.	yellow
4-dimethyl amino-benzhydrazide	pyridine-4-aldehyde	235° C.	yellow
4-dimethyl-amino-benzhydrazide	4-dimethylamino-benzaldehyde	279 to 280° C.	yellow
4-dimethylamino-benzhydrazide	9-methyl-carbazole-3-aldehyde	256 to 257° C.	colourless
4-dimethylamino-benzhydrazide	furfural	255 to 256° C.	colourless
4-dimethylamino benzhydrazide	anthracene-9-aldehyde	308° C.	yellow
benzhydrazide	anthracene-9-aldehyde	258 to 259° C.	yellow

1	2	3	4
pyridine-4-carboxylic acid hydrazide	4-nitro-benzaldehyde	278 to 279° C.	yellow
pyridine-4-carboxylic acid hydrazide	4,4'-bis-dimethylamino benzophenone	196 to 197° C.	yellow
pyridine-2-carboxylic acid hydrazide	4-dimethyl-amino-benzaldehyde	206 to 207° C.	yellow
pyridine-2-carboxylic acid hydrazide	4-diethylamino-benzaldehyde	127° C.	yellow
2-benzyl-amino-benz-hydrazide	4-dimethylamino-benzaldehyde	188° C.	yellow
4-diethyl-amino-benzhydrazide	4-diethylamino-benzaldehyde	182° C.	yellow
benzhydrazide	9,10-phenanthrene-quinone	192 to 193° C.	orange-coloured
pyridine-4-carboxylic acid hydrazide	9,10-phenanthrene-quinone	214° C.	orange-coloured
adipic acid dihydrazide	4-dimethylamino-benzaldehyde	293 to 296° C.	colourless
stearic acid hydrazide	4-dimethylamino-benzaldehyde	95 to 96° C.	pale yellow
stearic acid hydrazide	anthracene-9-aldehyde	149 to 150° C.	yellow
phenylacetic acid hydrazide	2-acetyl-9-ethyl-carbazole	173° C.	colourless
benzhydrazide	2-acetyl-9-ethyl-carbazole	184° C.	colourless
4-dimethylamino-benzhydrazide	2-acetyl-naphthalene	218° C.	pale yellow
4-dimethylamino-benzhydrazide	2-acetyl-fluorene	267 to 268° C.	yellow
phenyl-acetic acid hydrazide	4-dimethylamino-benzaldehyde	187° C.	colourless
phenyl-acetic acid hydrazide	anthracene-9-aldehyde	258° C.	yellow
adipic acid dihydrazide	2-acetyl-naphthalene	236 to 237° C.	colourless

1	2	3	4
4-dimethyl-amino-benzhydrazide	benzal-acetone	199 to 200° C.	pale yellow
formhydrazide	4-dimethylamino-benzaldehyde	161 to 162° C.	yellow
formhydrazide	anthracene-9-aldehyde	274 to 275° C.	yellow
formhydrazide	2-acetyl-9-ethyl-carbazole	220° C.	yellow

5 Certain of the compounds above referred may be dispersed with an insulating resin with an organic solvent and coated on to the base material or support by any known coating technique whereby solvent coatings are applied.

10 An aqueous solution of an insulating resin may be used as a solvent for selected compounds thus enabling the base material or support to be coated by the preferred aqueous coating method.

Various binders for the photoconductive material may be employed according to this invention as follows:

15 natural and synthetic resins, e.g. balsam resins, phenol resins modified with colophony and other resins of which colophony constitutes the major part, coumarone resins and indene resins and the substances covered by the collective term "synthetic lacquer resins", which includes processed natural substances such as cellulose ethers; polymers, e.g. the polyvinyl chlorides, polyvinyl acetates, polyvinyl acetals, polyvinyl alcohols, polyvinyl ethers, polyacrylic and polymethacrylic esters and polystyrene and polyisobutylene;

25 polycondensates, e.g. polyesters, such as phthalate resins, alkyd resins, malcate resins, maleic resins/colophony/mixed esters of higher alcohols, phenol-formaldehyde con-

densates, urea-formaldehyde resins, melamine-formaldehyde condensates, aldehyde resins, ketone resins, xylene-formaldehyde resins and polyamides;

polyadducts, such as polyurethane.

Suitable base materials or supports for use in the manufacture of the material of the present invention may be metal or glass plates, paper, or plates or foils of electrically conductive resins or plastics resins. Although glass is usually regarded as an insulating substance, it has been found to be sufficiently conductive for use as an electroconductive support.

45 Where a paper base material is used it may be desirable to pretreat it with, for example, an aqueous solution of methyl cellulose or an aqueous solution of polyvinyl alcohol to prevent penetration of the coating solution.

50 Sensitisers may be added to increase the spectral sensitivity of the photoconductive layer. The amount of sensitizer to be added to the photoconductive substance depends on the type of sensitizer and on the composition of the electrophotographic layer and it may vary within wide limits. For example good sensitising effects are obtained with an addition of 1% of Rhodamine B Extra. The most suitable sensitizers are chlorophyll and dyestuff compounds a number of which are given in the following table.

Dyestuff Group	Dyestuff Compound	Reference
		(Schultz' "Farbstoff-tabellen," 7th edn., Vol. 1 (1931))
Triarylmethane dyes	Brilliant Green	No. 760 (p. 314)
	Victoria Blue B	" 822 (p. 347)
	Methyl Violet	" 783 (p. 327)
	Crystal Violet	" 785 (p. 329)
	Acid Violet 6B	" 831 (p. 351)
Xanthene dyes: Rhodamines	Rhodamine B	No. 864 (p. 365)
	Rhodamine 6G	" 866 (p. 366)
	Rhodamine G extra	" 865 (p. 366)
	Sulphorhodamine B	" 863 (p. 364)
	Fast acid Eosin G	" 870 (p. 368)
Phthaleins	Eosin S	No. 883 (p. 375)
	Eosin A	" 881 (p. 374)
	Erythrosin	" 886 (p. 376)
	Phloxin	" 890 (p. 378)
	Rose Bengal	" 889 (p. 378)
	Fluorescein	" 880 (p. 373)
Thiazine dyes	Methylene blue	No.1038 (p. 449)
Acridine dyes	Acridine yellow	No. 901 (p. 383)
	Acridine orange	" 908 (p. 387)
	Trypaflavine	" 906 (p. 386)
Quinoline dyes	Pinacyanol	No. 924 (p. 396)
	Cryptocyanine	" 927 (p. 397)
Quinone dyestuffs		
Ketone dyestuffs	Alizarin	No. 1141 (p. 499)
	Alizarin red S	" 1145 (p. 502)
	Quinizarine	" 1148 (p. 504)
Cyanine dyes	Cyanine	" 921 (p. 394)

Following is a description by way of example of methods of carrying the invention into effect.

5

#### EXAMPLE 1.

A solution of the following was made up:—

- |   |        |
|---|--------|
| Benzaldhyde phenylhydrazone                             | 1 g.   |
| The ketone resin sold under the trade name Kunstharz Sk | 3 g.   |
| Methyl cellosolve (Registered Trade Mark)               | 15 ml. |

and coated by means of a plate whirler on to a metal plate and the coating dried.

- The thus coated plate was charged electrostatically negatively in the dark and exposed to a mercury arc lamp for 30 seconds through a film original.
- The latent electrostatic image thus formed was developed by application of a developer powder comprising a finely divided thermoplastic synthetic resinous material each particle of which was embodied with it carbon black,

the said finely divided material being mixed at about 2½% concentration with glass balls of about 300μ diameter and the image fixed.

25

#### EXAMPLE 2.

An aqueous solution of the following was made up:—

- |   |          |    |
|---|----------|----|
| Ortho-sulphobenzaldehyde phenylhydrazone              | 3 g.     | 30 |
| Urea formaldehyde Resin BC6 (50% soln.).              | 6ml.     |    |
| Water   | 24 ml.   |    |
| Sulphosalicylic acid and coated on to a metal support | 0.075 g. | 35 |

The coating was dried and cured for five minutes at 140° C. and processed as in Example 1.

#### EXAMPLE 3.

- |   |    |
|---|----|
| 0.5 g. of 4-diethylaminobenzylidene nicotinic acid hydrazide and 0.5 g. of ketone resin, e.g. | 40 |
|---|----|



the product commercially available under the trade name Kunstharz SK, were dissolved in 15 c.c. of glycol monomethylether and the solution applied to paper of which the surface had been pretreated against the penetration of organic solvents and it was dried. With the paper thus coated a direct image was produced by the electrophotographic process. The dry coating was provided by a corona discharge with a negative electric charge by means of a charging device maintained at approximately 6000 volts. It was then exposed under a positive master to the light of a high-pressure mercury lamp and powdered over with a developer in known manner. This developer consisted of toner and carrier. The toner was composed of a low melting point polystyrene, colophony and carbon-black, with or without an organic dyestuff additive such as spirit-soluble nigrosine, the components were melted together, ground and fractioned by screening. The most suitable fraction was that with a grain size of 20—60 $\mu$ . The toner, prepared in this way, was mixed with a carrier substance of such nature that the toner became triboelectrically charged with a charge that is the opposite of that produced on the paper, e.g. glass balls or iron filings. A positive image was produced which was fixed by slight heating. The ground of the paper was brightened by the substance applied as coating.

#### EXAMPLE 4.

The coating of the paper was carried out as described in Example 3 and coating provided with a positive charge by the corona discharge. After the paper foil had been exposed to light under a master, the image produced thereon was developed by powdering over with a developer, as described in Example 3, but as carrier glass balls covered with maleic acid resin were used. A very good, positive image of the master, rich in contrast, was obtained.

#### EXAMPLE 5.

0.5 g. of 4-dimethylaminobenzylidene-isonicotinic acid hydrazide and 0.5 g. of coumarone resin were dissolved in 15 c.c. of ethylene glycol monomethylether and the solution applied to a paper foil. After evaporation of the solvent an electrophotographic image was produced as described in Example 3.

#### EXAMPLE 6.

0.5 g. of 4-dimethylaminobenzylidene-benzhydrazide and 0.5 g. of phenol-formaldehyde resin were dissolved in 15 c.c. of ethylene glycol monomethylether. The application of the solution to a paper foil and the preparation of the electrophotographic image were as described in Example 3.

#### EXAMPLE 7.

The procedure described in Example 3 was followed, but for the coating of the paper the solution of 0.5 g. of 4-dimethylaminobenzylidene nicotinic hydrazide and 0.5 g. of zinc resin, e.g. the product marketed under the trade name Erkazit Zinkharz 165, in 15 c.c.

of ethylene glycol monomethylether was used.

#### EXAMPLE 8.

0.5 g. of 4-diethylaminobenzylidene-benzhydrazide and 0.5 g. of ketone resin, e.g. the product marketed under the trade name Kunstharz S.K., were dissolved in 15 c.c. of ethylene glycol monomethylether. This solution was applied to transparent paper, the surface of which had been pretreated against the penetration of organic solvents, and it was then dried. On this coated transparent paper images were produced by the electrophotographic process which were fixed by heating or by treatment with trichlorethylene vapours. They were then used as intermediate originals for further duplication, e.g. for copying on diazo paper.

#### EXAMPLE 9.

0.5 g. of 4-methoxy-benzylidene-benzhydrazide and 0.5 g. of non-hydrolysed ketone/aldehyde condensation resin, e.g. the product marketed under the trade name Kunstharz AP, were dissolved in 15 ml. of ethylene glycol monomethylether and applied to a superficially roughened aluminium surface. After the solvent had evaporated, a coating was left that was firmly adherent to the surface of the foil. With this coated foil the procedure described in Example 3 was followed and a positive image was obtained on the aluminium surface once the powder image had been fixed. This image can be converted into a printing plate if the aluminium foil is wiped over with 50% acetic acid or 60% ethanol, rinsed down with water and then inked up with 1% phosphoric acid and greasy ink. A positive printing plate is obtained which can be set up in an offset machine and used for printing.

#### EXAMPLE 10.

A mixture consisting of:—

0.1 g. of 4-dimethylaminobenzylidene-2-toluy-hydrazide  
0.1 g. of 2-chloro-4-dimethylaminobenzylideneisonicotinic acid hydrazide.  
0.1 g. of furfurylidene-(2)-4<sup>1</sup>-dimethylamino-benzhydrazide  
was dissolved in 15 c.c. of ethylene glycol monomethylether. The solution was applied to an aluminium foil and then dried to form a coating that was firmly adherent to the metal. The preparation of an electrophotographic image was carried out in manner known *per se*.

#### EXAMPLE 11.

0.5 g. of 4-diethylaminobenzylidene nicotinic acid hydrazide, 0.5 g. of ketone resin, e.g. the product already mentioned in Example 3 marketed under the trade name Kunstharz SK, and 5 mg. of Rhodamine B extra were dissolved in 15 c.c. of ethylene glycol monomethylether. This solution was applied to paper of which the surface had been treated against the penetration of organic solvents and it was then dried. With this material an electrophotographic image was produced by the method described in Example 3 but instead

of the high-pressure mercury lamp a 100-watt incandescent bulb was used.

#### EXAMPLE 12.

8 g. of ketone resin, e.g. the product commercially available under the trade name Kunstharz EM, were dissolved in 120 c.c. of ethanol. To this solution 8 g. of anthracene-9-aldehyde-isonicotinoyl hydrazone were added. The suspension thus produced was very finely ground in a ball mill. This suspension was then coated upon paper that had been pretreated against the penetration of organic solvents and was then dried. Electrophotographic images were prepared with the coated paper by the process described in Example 3.

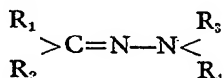
If instead of the 8 g. of anthracene-9-aldehyde - isonicotinoyl hydrazone the same quantity of N,N'-bis - (4 - dimethylamino-benzylidene) - adipic acid dihydrazide is used, a similar result is obtained.

#### EXAMPLE 13.

10 g. of chlorinated polyvinyl chloride were dissolved in 100 c.c. of methyl ethyl ketone. To this solution 10 g. of 4-dimethylamino-benzylidenebenzhydrazide dissolved in 50 c.c. of toluene were added. With the mixture thus obtained paper was coated by means of a hopper device. When the coated solution had dried to a firmly adherent homogenous layer, direct image were produced electrophotographically on this paper by the method described in Example 3. The electrostatic charge was given up very quickly in the places in which the photoconductive coating was struck by the light. Light-sensitivity was good. Images rich in contrast were obtained episcopically from double sided masters.

#### WHAT WE CLAIM IS:—

1. An electrophotographic material consisting of a conductive support and a photoconductive insulating layer adherent thereto, which layer comprises an organic compound of general formula



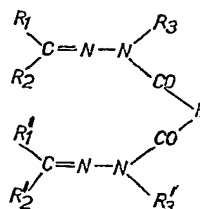
in which  $R_1$ ,  $R_2$  represent hydrogen, alkyl, aralkyl, acyl, aroyl, cycloalkyl, or univalent radicals of aromatic or substituted aromatic carbocyclic or heterocyclic ring systems and wherein  $R_1$  and  $R_2$  may join to form a ring, and in which  $R_3$ ,  $R_4$  represent hydrogen, alkyl, aralkyl, aroyl, cycloalkyl, or univalent radicals of aromatic or substituted aromatic carbocyclic or heterocyclic ring systems and wherein  $R_3$  or  $R_4$  may represent acyl.

2. A material as claimed in claim 1, wherein the organic compound is a hydrazone of an aldehyde.

3. A material as claimed in either of the preceding claims, wherein the organic compound is a phenylhydrazone derivative of an aldehyde or ketone.

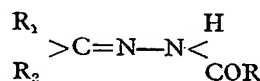
4. A material as claimed in any one of the preceding claims, wherein the organic compound is benzaldehyde phenylhydrazone or *o*-sulphobenzaldehyde phenylhydrazone.

5. A material as claimed in claim 1, wherein the organic compound is formed by the reaction of a dihydrazide with an aldehyde or ketone and has the general formula:—



in which  $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_4$  represent hydrogen, alkyl, aralkyl, acyl, aroyl, cycloalkyl, or univalent radicals of aromatic or substituted aromatic carbocyclic or heterocyclic ring systems wherein  $R_1$ ,  $R_2$  and/or  $R_3$ ,  $R_4$  may join to form a ring, in which R is alkylene and in which  $R_3$ ,  $R_4$  represent hydrogen, alkyl, aralkyl, aroyl, cycloalkyl, or univalent radicals of aromatic or substituted aromatic carbocyclic or heterocyclic ring systems.

6. A material as claimed in any one of claims 1, 2 or 5, wherein the organic compound is an acyl hydrazone having the general formula



in which R represents H, alkyl, aralkyl, aryl, substituted aryl, or a heterocyclic residue of aromatic character,  $R_1$  represents H, alkyl, aryl or substituted aryl,  $R_2$  represents aralkyl, aryl, substituted aryl or a heterocyclic residue of aromatic character in which  $R_1$  and  $R_2$  may join to form a ring.

7. A material as claimed in any one of the preceding claims wherein the insulating layer also contains sensitizers.

8. A process for the preparation of an electrophotographic material substantially as herein described with reference to any one of the specific examples.

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